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ScienceDirect
Physics Procedia 12 (2011) 166–173
Physics Procedia

LiM 2011

Experimental Studies on Laser-based Hot-melt Bonding of thermosetting Composites and Thermoplastics

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Abstract

This paper presents experimental results of joining carbon fiber (CF) reinforced thermosetting composites (TSC) to thermoplastics (TP) by means of laser-based hot-melt bonding. First of all the influence of different laser systems ($\lambda = 355$ nm and $\lambda = 1064$ nm) on the ablation of the thermosetting matrix (epoxy) with preferably little damage to the CF is analyzed by means of microscopy. Afterwards the laser-based joining process of TSC and TP is carried out. Finally the joining connections are characterized by tensile shear tests. Thereby the influence of the surface treatment and the used thermoplastic (PC/ABS, PA66 or PA66-GF30) on the tensile shear strength is investigated.

Keywords: laser joining; hot-melt bonding; lightweight design; thermosetting composite; carbon fiber reinforced plastic; thermoplastic

1. Introduction

In recent years lightweight design by means of carbon fiber reinforced plastic (CFRP) became a focus in the construction of automobiles, wind power plants and aircrafts due to the scarcity of raw materials and the increasing environmental awareness [1]. As matrix which impregnates the fibers (e.g. carbon or glass fibers) mostly thermosetting resins (e.g. epoxy or polyester resins) are used because of their better wetting of the fibers and their simple processing in comparison to thermoplastic matrices. 40 years ago the mass fraction of carbon fiber (CF) structures in aircrafts like the Airbus A300 was only 5% whereas an A380 built 2005 already consists of 22% CFRP. The new status symbol from Boeing, the so-called Dreamliner (Boeing 787), has already a CF mass fraction of about 60% [2]. In the past few years in the automotive industry a lot of steel components are substituted by aluminum parts to reduce the car weight for lower fuel consumption and emission. The modern lightweight design by means of CFRP allows a further reduction of weight whereby the technological progress of CFRP is still increasing (see Figure 1). As a result of their versatile applicability and their outstanding technical properties CFRP will be used in future even more in various industrial products. However in all industries, the lightweight potential and product-specific requirements can only be fulfilled by an optimal and intelligent integration of different

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doi:10.1016/j.phpro.2011.03.021

materials and suitable joining technologies [3]. According to the current state of the art the joining of thermosetting composite (TSC) to thermoplastic (TP) is limited by the available joining techniques (see Figure 2).

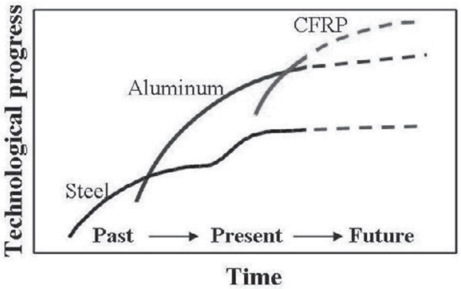

Figure 1: Comparison between the technological progress of steel, aluminum and CFRP [according to 4]

Thermosetting plastics can not be molten because their macromolecules are cross-linked so that welding is only possible for thermoplastic matrix composites (TPC) [5]. Dissimilar materials such as thermoplastic and thermosetting plastics can be joined by means of bonding. However a major disadvantage of bonding is the time-consuming and complex surface preparation and the long curing time which can vary by product and type of adhesive between several minutes and several hours [6]. As an alternative to bonding integral design can be used. Thereby the TSC is coated with a thermoplastic layer to make the composite weldable. The coating can be realized by a thermoplastic hybrid interlayer or a thermoplastic film co-cure. Both processes are directly linked to the manufacturing process of the TSC. A subsequent application of the TP layer on the TSC after the curing process is not possible and makes these techniques inflexible [7]. For a mechanical linkage with rivets or bolts holes have to be drilled into the composite whereby the fibers are cut through and the flux of force in the CFRP is significantly influenced. This has to be considered in the component design. Besides the fact that mechanical linkage is not “fiber-friendly” the joining elements limit the lightweight design and the visual appearance [8].

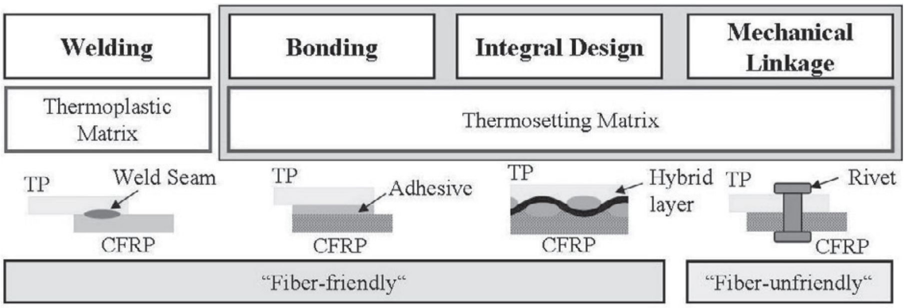

Figure 2: Current joining techniques for CFRP and TP

2. Laser-based hot-melt bonding of thermosetting composites and thermoplastics

Motivated by the described deficits of available joining techniques an innovative approach for joining thermosetting composites to thermoplastics by means of laser-based hot-melt bonding is presented in this paper. The new joining technique can be divided into (see also Figure 3):

- the laser-based surface treatment of the thermosetting composite and
- the laser-based joining process of the thermosetting composite and the thermoplastic.

The laser-based surface treatment is used to ablate the thermosetting matrix from the fibers to enlarge the effective joining area. This laser ablation causes a rough surface with micro undercuts (see Figure 3 a) which is important for the interlocking between the joining partners that takes place in the following joining process (see

Figure 3 b). After the pretreatment the joining partners are arranged in overlap and fixed under pressure whereby the thermoplastic is positioned over the pretreated CFRP. The joining process is similar to transmission laser welding and requires a laser transmissive thermoplastic material. During the joining process the laser radiation is absorbed primarily by the carbon fibers of the lower joining partner (here: CFRP) whereby in contrast to conventional transmission laser welding no melt arises because the fibers and the thermosetting matrix can not be molten. However the absorbed laser beam heats the thermosetting CFRP. By heat transfer the TP is molten and acts as a hot-melt adhesive which wets the fibers and leads to an interlocking of the joining partners. After cooling down of the thermoplastic melt (hot-melt adhesive) the TP and the TSC are permanently joined together.

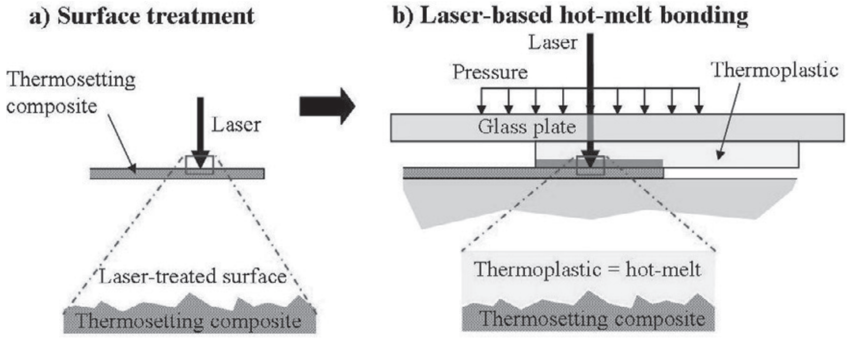

Figure 3: (a) Laser-based surface treatment of the thermosetting composite; (b) Laser-based hot-melt bonding

3. Experimental

In this paper experimental studies are conducted to the laser surface treatment of carbon fiber (CF) reinforced thermosetting composites (TSC) to verify the influence of laser-treated surface on the laser-based hot-melt bonding. Additionally the influence of the used thermoplastic respectively hot-melt adhesive on the joining connection is analyzed.

3.1. Laser-based surface treatment

As specimens for the laser pretreatment CF reinforced TSC with epoxy resin ($l \cdot w \cdot h$ = 70 mm · 20 mm · 0,7 mm) are used. The specimens are laser-structured in the overlap area A_{max} ($A_{max} = l_o \cdot w$ = 7 mm · 20 mm). To ablate the epoxy matrix two different laser systems with UV (λ = 355 nm) and IR (λ = 1064 nm) radiation are utilized (see Table 1). The aim of the performed experiments is to provide information about wavelength or laser parameters which are suitable to realize an ablation of the epoxy matrix with preferably little damage to the carbon fibers. Another target is to clean the surface totally without matrix residues. The pretreated surfaces are analyzed by microscopy (Keyence VHX – 100K) and visually analyzed according to the described targets.

Table 1: Properties and parameters of the used laser systems

Property / Laser	Nd:YAG laser	Frequency-tripled Nd:YVO_4 laser
Wavelength λ [nm]	1064	355
Focus diameter d [µm]	40	50
Max. laser power $P_{max.}$ [W]	10	4
Hatch distance h [mm]	0,1	0,1
Type	VMC3; TRUMPF	Navigator II YHP 40-355QWA; Spectra Physics

3.2. Laser-based hot-melt bonding

Besides the “fiber-friendly” ablation it is important that a strong joint can be produced by laser-based hot-melt bonding. The laser-based hot-melt bonding as described in chapter 2 (see Figure 3b) is performed by a diode laser (λ = 940 nm, P_{max} = 50 W) with an elliptical focus diameter of about 1 mm. The specimens are fixed under constant pressure and the overlap area A_{max} is scanned by the laser multiple times so that the thermoplastic is heated quasi-simultaneously. For the experiments pretreated CFRP (see Chapter 3.1) and different thermoplastics ($l \cdot w \cdot h$ = 70 mm x 20 mm x 2 mm, see Table 2) are used.

Table 2: Properties of used thermoplastics

Property / Material	PC/ABS	PA66	PA66-GF30
Structure	Amorphous	Crystalline	Crystalline
Filler	None	None	30 wt.% glass fibers
Melting temperature [°C]	140	260	260
Yield stess [MPa]	40	85	170

The joining connections of the fabricated specimens are characterized by microscopy (Keyence VHX – 100K) and tensile shear tests according to DIN EN 1465. The tests are performed on a Zwick/Roell machine (used parameters see Table 3). The tensile shear strength τ can be calculated by Equ. 1 whereby F_{max} is the maximum force and A_{max} the overlap area which is defined by the overlap length l_o of the joining partners and the width w of the specimens:

$$\tau = \frac{F_{max}}{A_{max}} = \frac{F_{max}}{w \cdot l_o} \tag{1}$$

Table 3: Parameters of the tensile shear test

Parameter	Value
Overlap area A_{max} [mm^2]	140
Feed rate v_F [mm/min]	5
Clamping length l_c [mm]	50

4. Experimental results and discussion

4.1. Laser-based surface treatment

The performed experiments show that laser treatment with UV and IR radiation cause wavelength-dependent results. First of all the results for the laser treatment of the CFRP with IR (λ = 1064 nm) radiation are reported. The used matrix is highly transmissive for the Nd:YAG laser so that the carbon fibers primarily absorb the radiation. Besides the fibers have a higher heat conductivity and evaporation temperature than the epoxy matrix. This fact causes that the matrix is heated up on its evaporation temperature by heat transfer. Figure 4 presents results of the pretreatment with a Nd:YAG laser (laser-treated 2 times) whereby the laser power is varied from 1,7 W up to 2,7 W.

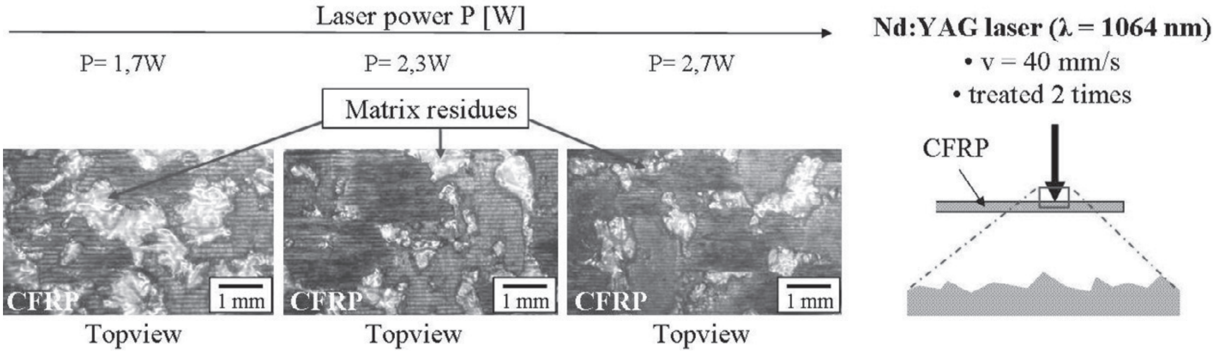

Figure 4: Topview of laser-treated (λ = 1064 nm) CFRP in dependence of the laser power

The experiments show that a total ablation of the epoxy resin from the surface is difficult by means of a Nd:YAG laser (see Figure 4). The only chance to realize a holohedral ablation of the resin with IR radiation is to repeat the laser treatment several times. The results of the multiple irradiations are documented in Figure 5.

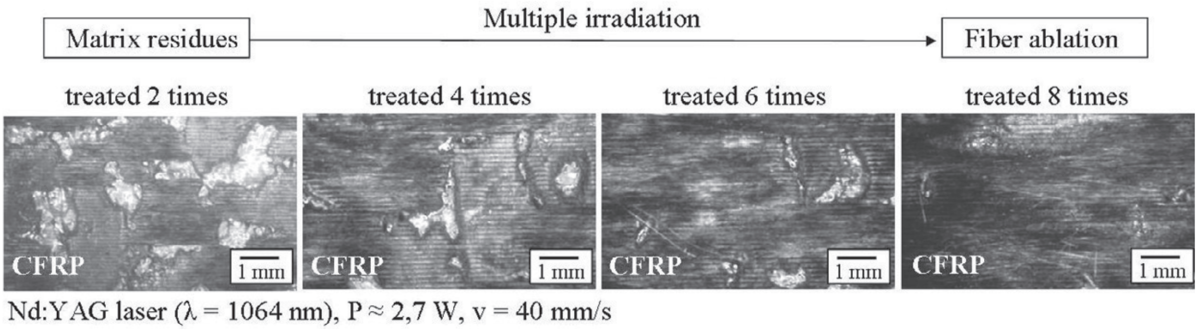

Figure 5: Topview of laser-treated (λ = 1064 nm) CFRP in dependence of multiple irradiation (treated 2 till 8 times)

However a multiple irradiation (e.g. 8 times, see Figure 5 right) of the surface causes on the one hand the ablation of the matrix but one the other hand although the ablation of the CF which is unwanted. In contrast to that the experimental studies with the UV laser (λ = 355 nm) demonstrate that a holohedral ablation of the epoxy resin with hardly no damage of the fibers is possible. The results of the laser treatment of both laser systems can be compared in Figure 6.

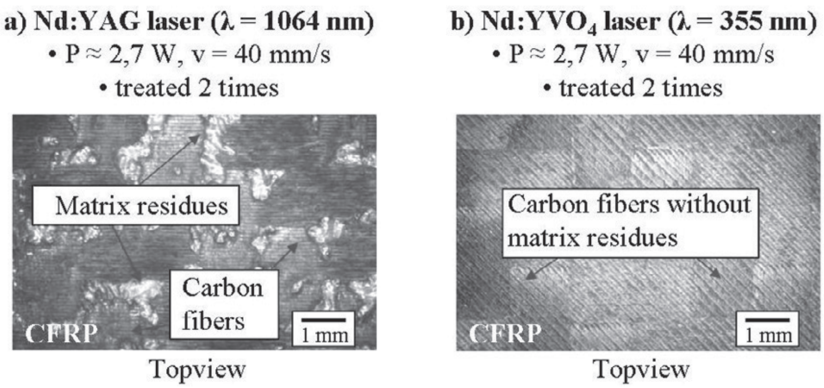

Figure 6: (a) Topview of IR laser-treated (λ = 1064 nm) CFRP; (b) Topview of UV laser-treated (λ = 355 nm) CFRP

The different results can be explained by the fact that the epoxy resin like most plastics has a higher absorption of UV than IR radiation which favors the ablation of the matrix by means of frequency-tripled $Nd:YVO_4$ laser. Similar results for UV ablation of CFRPs can be looked up in [9]. The photon energy of the UV laser is high enough to directly break the chemical bonds of the matrix whereby for an IR laser the ablation of the epoxy is caused by heat which is absorbed by the carbon fibers. In addition, the micrographs in Figure 6 a, b present that more fibers are cut through by the IR than by the UV laser (see also 7 a, b).

4.2. Laser-based hot-melt bonding

After the laser pretreatment (see Figure 6 a, b) the CFRP and the TP are joined by laser-based hot-melt bonding (see chapter 3.2). Micrographs of the joints (PC/ABS + CFRP) are shown in Figure 7. There you can see that still some matrix residues are on the Nd:YAG laser-treated surface (see Figure 7 a) whereas the UV laser-treated surface is totally cleaned (see Figure 7b). Besides the matrix residues there are broken fibers in the joining zone that are caused by the IR pretreatment (see Figure 7 a).

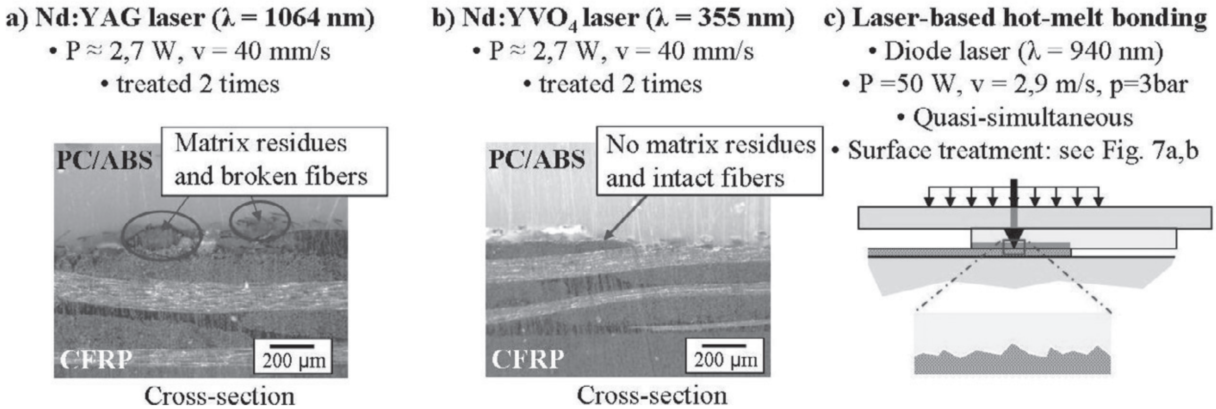

Figure 7: (a) Micrograph of the cross-section of the joint (PC/ABS + CFRP, IR laser-treated); (b) Micrograph of the cross-section of the joint (PC/ABS + CFRP, UV laser-treated); (c) Joining parameters for laser-based hot-melt bonding

To not only qualitatively assess the surface treatment on the basis of micrographs tensile shear tests are carried out and their results are shown in Figure 8. Similar tensile shear strengths are reached for CFRP treated with IR and UV lasers and subsequently joined to PC/ABS by means of laser-based hot-melt bonding (see Fig 8).

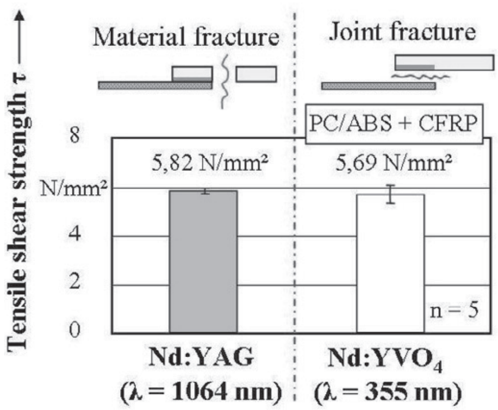

Figure 8: Tensile shear strengths of the IR and UV laser-pretreated joints (PC/ABS + CFRP) (parameters: see Figure 7 a,b,c)

However the failure behaviors of the used specimens are different (see Figure 8 top). All CFRP treated with the UV laser crack at the joint whereas this area for IR laser-treated specimens stays intact and a material fracture of the PC/ABS occurs. This leads to the conclusion that a “fiber-friendly” ablation of the thermosetting matrix can be realized by UV laser treatment. However IR laser-treatment should be used for high tensile shear strength. The broken fibers and the matrix residues which are the result of the IR laser treatment (see Figure 6a and 7a) increase the adhesive surface and cause a better interlocking between the thermoplastic melt and the CFRP. In this case a compromise between “fiber-friendly” ablation and high shear strength has to be made.

Besides the surface treatment the influence of the used hot-melt adhesive is investigated in this paper. In Figure 8 and 9a it can clearly be seen that the tensile shear strength is highly dependent on the used thermoplastic. The substitution of PC/ABS (see Figure 8) with PA66 or PA66-GF30 (see Figure 9 a) leads to an significant increase of the tensile shear strength from about 6 N/mm^2 up to 10 till 12 N/mm^2. The reinforcing fibers of the PA66-GF30 additionally enhance the mechanical interlocking between the joining partners in contrast to unfilled thermoplastics (see Figure 9 b). However in contrast to the unfilled thermoplastic PA66-GF30 cracks at the joint (see Figure 9 a top and Figure 9 c) which leads to the conclusion that the maximum tensile shear strength of the joint for the used pretreatment and the used joining parameters is reached.

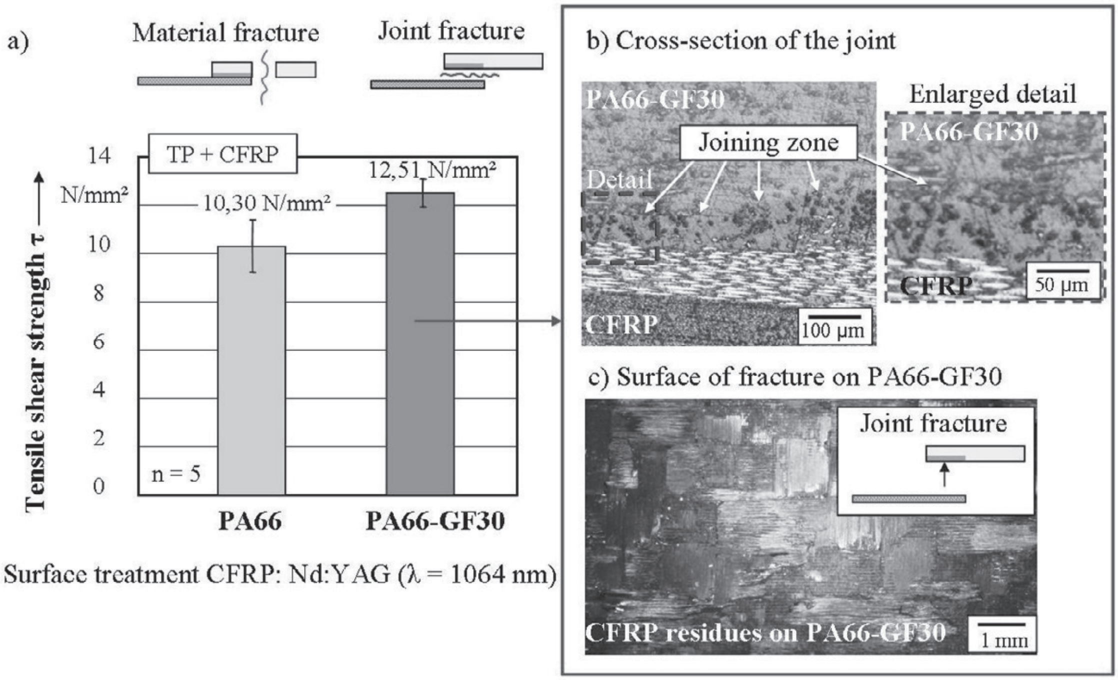

Figure 9: (a) Tensile shear strengths of joints in dependence of the used thermoplastic (PA66+ CFRP; PA66-GF30 + CFRP); (b) Micrograph of the cross-section of the joint (PA66-GF30 + CFRP, IR laser-treated); (c) Micrograph of the surface of the fracture on PA66-GF30

5. Conclusion and outlook

In the future lightweight potential and product-specific requirements can only be fulfilled by an optimal and intelligent integration of different materials. Therefore suitable joining technologies are necessary. However according to the current state of the art the joining of thermosetting composite (TSC) to thermoplastic (TP) is limited by the available joining techniques. In this paper a new approach for joining TSC and TP by the use of laser-based hot melt bonding is presented. The new joining technique includes the laser-based surface treatment of the TSC and the laser-based joining process of the TSC and the TP.

For this purpose carbon fiber (CF) reinforced thermosetting composites are treated with different laser systems (see Table 1) to analyze the influence of the laser treatment on the laser-based hot-melt bonding (see Chapter 3.1, 4.1). Besides the influence of the used thermoplastic on the joining connection is determined (see Chapter 3.2, 4.2).

The performed laser pretreatment of the thermosetting CFRP show that UV radiation (λ = 355 nm) ablates the epoxy matrix better than IR radiation (λ = 1064 nm). This can be explained by the different beam-material-interaction. However the experiments also show that “fiber-friendly” ablation of the thermosetting matrix is not directly connected to high tensile shear strength of the joint. In contrast to the UV treatment the IR radiation breaks some carbon fibers and causes matrix residues on the surface which lead to undercuts and enlarge the effective joining area. All in all a compromise between “fiber-friendly” ablation and high shear strength has to be made.

Besides it is shown that for a strong joint the used thermoplastic is very important. Furthermore it is investigated that filled hot-melt adhesives (e.g. glass fibers) enable higher tensile shear strengths because of a additional mechanical interlocking between the joining partners.

All in all the reported experiments demonstrate that laser-based hot-melt bonding is a suitable solution for joining TSC to thermoplastic TP. Because of the fact that laser-based hot-melt bonding is a new joining technique there are still some unanswered issues. Current investigations are focused on the fundamental question if the laser pretreatment is really necessary or if this process step can be substituted for example by optimized laser joining strategies. Additionally further experiments with other filled and unfilled thermoplastics like for example PC or PP are planned.

Acknowledgements

The authors acknowledge support from CrossLink Faserverbundtechnik GmbH & Co.KG and thank the Federal Ministry of Economics and Technology for funding the ZIM-project “Laser-based hot-melt bonding of TSC and TP - Research and realization of an innovative joining technique for TSC”.

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